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DETAILED DESCRIPTION

[Detailed Description of the Invention]
A direct type methanol supply type fuel cell and its system This application is U.S. patent application 08th / continuation-in-part application of No. 478,001 for which it applied on June 7, 1995 of U.S. patent application 08th for which it applied on October 12, 1993 / continuation application of No. 135,007.
The invention of ****** of an invention relates to the direct supply type methanol fuel cell by which it was improved for the system which operates without using an acid electrolyte or a reformer.
The transportation vehicles which operate with a background and outline gasoline power type internal-combustion engine are becoming a source of many environmental problems. The product which comes out of an internal-combustion engine produces the problem to which smog and other exhaust gas related, for example. By various measures against pollution control, the exhaust gas component which a certain kind does not have is made into the minimum. However, the process of combustion generates a certain exhaust gas intrinsically. However, even if it can make exhaust gas environment-friendly extensively, it still depends for the internal-combustion engine which uses gasoline as a base on the fossil fuel of non-reproducibility.
Many groups have explored the suitable solution of these energy problems.
The fuel cell is mentioned as solution with one possibility. A fuel cell reacts chemically using the energy from a refreshable fuel material. For example, methanol is a completely refreshable supply source. A fuel cell uses oxidation/reduction reaction instead of a combustion reaction. Typically, most of the final product acquired from the reaction of this fuel cell are carbon dioxide and water.
Some conventional methanol fuel cells used the "reformer", in order to change methanol into H_2 gas used with a fuel cell. The methanol fuel cell used the electrolyte of strong acid. The

artificers of this invention proposed the art of operating a fuel cell directly from methanol, and what is called a direct supply type fuel cell, without using an acid electrolyte for the beginning. The theme matter of this improvement is our United States patent ****.
It is alike, and is indicated and the indication matter of the patent concerned is used for a grade required for a suitable understanding as reference at this specification. Since this is achievements of the artificers, us, of course, it does not recognize here that this patent constitutes the advanced technology over this invention.
The theme of this invention explains the further improvement of such a direct supply type fuel cell. Based on the further research achievements of the artificers about this concept, the various improvement to the structure of a fuel cell itself is indicated here. Preparation by which the electrode which improves an operation of an electrode was improved is included in these improvement matters.
The improvement catalyst which raises the efficiency of methanol generation is included in the operation of an electrode. An expensive platinum catalyst is used for a fuel cell. The electrode method of preparation provided here defines the art which can make insufficient [make decrease and] the necessity of using a platinum catalyst.
The art which forms a cathode terminal is also indicated here. Such art makes the optimal an operation of the cathode used with incompressible air. By this, ambient air temperature and atmospheric pressure can be permitted now in a reduction mechanism, and the efficiency of a fuel cell can be improved further.
The art of electrode formation is also explained and the art of preparing membranous conditions is also included in it. Formation of the desirable membrane electrode assembly is also defined especially.
This invention also defines the design of the flow field which makes it easy to supply liquid fuel to a catalyst again.
This fuel cell system needs to be used in a final product after all. This final product may be an internal-combustion engine, or may be much easy electronic equipment like radio. Any products which operate electrically can be operated based on the electric power generated from these fuel cells. The artificers of this invention found out a certain art of improving many of these problems that may happen, by other methods while improving this operation.
the art of this invention -- "the system operation" was enabled by explaining the art of operating this fuel cell as some whole systems.
The sensor for measuring methanol concentration and other important parameters is contained in these system technologies. This artificer understands the thing for which various sensors for detecting various parameters will be required. This artificer was not able to find a commercial sensor. The method of transforming into this invention the art of using with this fuel cell, for formation of a sensor is indicated. Using the art of this fuel cell, this sensor maintains high

reliability and operates.
Another art defines formation of a unipolarity cell.
easy **** of a drawing -- this invention -- these -- it reaches and other fields are explained in detail below with reference to an accompanying drawing.
Drawing 1 and 2 show the fundamental fuel cell by this invention.
Drawing 3 shows the dish for desiccation used in order to dry the carbon paper sheet which encoded Teflon.
Drawing 4 shows the fundamental platinum sputter device of this invention.
Drawing 5 shows the fundamental flow field device which takes like the 1st operative condition as for this invention.
Drawing 6 shows the sectional view of the desirable flow field cut along the line 6-6 of drawing 5.
Drawing 7 shows the 1st embodiment of the by plate structure of this invention.
Drawing 8 shows the 2nd embodiment of by plate structure.
Drawing 9 shows the system operation of a direct type methanol field fuel cell.
Drawing 10 shows how the concept of the fuel cell mentioned above in order to use by a methanol sensor should be changed.
Drawing 11 shows the relation of the methanol concentration and current in this invention.
Drawing 12 shows the fuel cell of the molecular sieve which gave the stage to the class for methanol by this invention.
Drawing 13 shows the figure to which the 1st about the way in the single electrode of the fuel cell of this invention was expanded.
Drawing 14 shows package-ization of the way in this single electrode.
Drawing 15 is an enlarged drawing showing the 2nd embodiment of the way in a single electrode.
Drawing 16 shows how the way in this single electrode is assembled to an operation system.
Drawing 17 shows another expansion layout of the assembly of the way in a single electrode.
a desirable operative condition -- explanation [like] -- the fluid distribution system indicated to the patent which made reference by our above -- an anode top -- a platinum-ruthenium catalyst -- the platinum catalyst is used on the cathode. perfluoro-sulfone membrane -- Nafion (Nafion) 117 of DuPont is preferably used as a polymer electrolyte film.
An important thing is that neither any acid electrolytes nor a reformer needed to use it, and this system operated. The various characteristics which various materials have were changed and this improvement was made.
By improving hydrophilic nature for an anode electrode using an ionomer additive agent, the accessibility of the methanol aqueous solution on the surface of an anode has been improved. The option which improves the hydrophilic nature of an anode was using the electrolyte

containing superacid ("C8 acid").
For example, since those molecular sizes of another methanol derivative fuel like trimethoxy methane "TMM" are large, the mobility (crossover) of fuel becomes small for other characteristics.
Drawing 1 shows the fluid supply organicity fuel cell 10 which has the housing 12, the anode 14, the cathode 16, and the solid polymer proton conductivity cation exchange electrolyte membrane 18. As explained much more in detail below, the anode 14, the cathode 16, and the solid polymer electrolyte film 18 are desirable single multilayer composite-construction objects. Here, a membrane electrode assembly is called.
The pump 20 is formed in order to carry out pumping of the solution of organic fuel and water into the anode room 22 of the housing 12. The mixture of the organic fuel and water is recycled through the recirculation system which has the methanol tank 19 which is taken out from the exit port 23 and explained below with reference to drawing 2. The carbon dioxide generated at the anode room is emitted through the port 24 in the tank 19. The compressor 26 of oxygen or air is formed in order to supply oxygen or air into the cathode room 28 in the housing 12.
Drawing 2 explained below shows the fuel cell system incorporating the stack of each fuel cell containing a recirculation system. The following detailed explanation of the fuel cell of drawing 1 mainly performs the structure and the function of the anode 14, the cathode 16, and the film 18 as a center.
In advance of use, the anode room 22 is filled with the mixture of organic fuel and water, and fills the cathode room 28 with air and oxygen. Organic fuel passes and circulates through the anode 14 during an operation, on the other hand, pumping of oxygen or the air is carried out into the cathode room 28, and it passes and circulates through the cathode 16. If electric load (not shown) is connected with the anode 14 between the cathodes 16, electric oxidation of organic fuel will take place with the anode 14, and electric reduction of oxygen will take place with the cathode 16. When such a different reaction occurs with an anode and a cathode, voltage difference arises in these 2 inter-electrode. The electron generated by electric oxidation with the anode 14 conducts through external load (not shown), and, finally is caught with the cathode 16. The film electrolyte 18 is crossed directly, the hydrogen ion, i.e., the proton, which were generated with the anode 14, and it is conveyed to the cathode 16. Therefore, the flow of current is maintained with the electron which passes the flow and external load of the ion which passes a cell.
As mentioned above, the anode 14, the cathode 16, and the film 18 form one composite layer structure. In a desirable embodiment, the film 18 is formed by Nafion (trademark) of perfluoro-** proton-exchange-membrane material. Nafion (trademark) is a copolymer of tetrafluoroethylene and perfluoro-vinyl ether sulfonic acid. Other charges of a film material can be used. For example, the film of change perfluoro-** sulfonic acid polymer, poly hydrocarbon

sulfonic acid, and the complex of two or more sorts of proton exchange membrane can be used.
From the particles of a platinum ruthenium alloy, it is made to distribute on carbon of the large surface area as detailed metal powder, i.e., "a structure where it does not support", and the anode 14 is got blocked, is "support structure" and is formed. Material like Vulcan XC-72A which an American Cabot Inc. company provides may be sufficient as carbon of large surface area. In order to perform an electrical link with the particles of the electrocatalysis, the backing (not shown) of a carbon fiber sheet is used. A commercial Toray (trademark) paper is used as an electrode backing sheet. The alloy electrocatalysis of the support structure on the backing of a Toray (trademark) paper can obtain from E-Tek of the U.S. Massachusetts state hula MINGAMU whereabouts, and an Inc. company. Or an anode can be manufactured by the chemical method which joins together with a Teflon (trademark) binding material, and sprinkles both the electrocatalysis of the structure where it does not support, and the electrocatalysis of support structure, on Toray (trademark) paper backing. How to manufacture an electrocatalysis electrode without spending many hours efficiently is explained in detail below. The second metal can use the alloy of the platinum base which is tin, iridium, osmium, or a rhenium instead of a platinum ruthenium. Generally, selection of an alloy is performed depending on the fuel used for a fuel cell. A platinum ruthenium is preferred in order to carry out electric oxidation of the methanol. In the case of a platinum ruthenium, the quantity of the alloy particles added in an electrocatalysis layer has the preferred range of 0.5 - 4.0 mg/cm^2. Electric oxidation with the more efficient higher one is realized rather than an addition level is low.
The cathode 16 is a gas diffusion electrode.
Platinum particles are joint ******** to one field of the film 18.
As for the anode 14 of the film 18, it is [the cathode 16] preferred to form with platinum of the structure combined with the field of the opposite hand where it does not support, or support structure. The platinum material of the support structure which can be obtained from the platinum black (fuel cell grade) of the structure where it does not support which can be obtained from an American Johnson Matthey Inc. company, or an American E-Tek Inc. company fits cathodes. It is preferred like [in the case of an anode] to carry the metal particles for cathodes on the carbonaceous charge of a backing material. The quantity which carries out load of the electrocatalysis particles on carbon backing is the range of 0.5 - 4.0 mg/cm^2 preferably. The backing of an electrocatalysis alloy and carbon fiber has given hydrophobicity required to remove efficiently the water which contained 10 to 50% of the weight of Teflon (trademark), and generated the three-phase zone, and was generated by electric reduction of oxygen.
During an operation, a density range passes the anode 14 in the anode room 22, and

circulates the mixture (neither an acid electrolyte nor an alkaline electrolyte is contained) of the fuel and water which are 0.5-3.0 mol/l. Preferably, the rate of flow of the range for 10 - 500mL/is used. When the mixture of fuel and water passes and circulates through the anode 14, in the case of an illustration methanol cell, the following electrochemical reaction occurs and electrons are emitted.

Anode: $CH_3OH+H_2O->CO_2+6H^++6e^-$ (1)

The carbon dioxide generated at the above-mentioned reaction is taken out through the exit 23 with the solution of fuel and water, and is separated from the solution by a gas liquid separation device (with reference to drawing 2, it explains below). Subsequently the solution of fuel and water is recycled into a cell with the pump 20.

Other electrochemical reaction which performs electric reduction of oxygen which catches an electron simultaneously with the electrochemical reaction shown by the above-mentioned formula (1) occurs with the cathode 16, and is expressed with a following formula.

Cathode: $O_2+4H^++4e^-$ -> H_2O (2)

By each electrode reaction expressed with the formula (1) and (2), the whole reaction expressed with a following formula is performed about an illustration methanol fuel cell.

Cell: $CH_3OH+1.5O_2->CO_2+2H_2O$ (3)

With the fuel of concentration high enough, current density higher than 500 mA/cm^2 is maintainable. However, in such concentration, the movement speed (crossover rate) of the fuel which crosses the film 18 and faces to the cathode 16 increases even to such an extent that the efficiency and electrical performance of the fuel cell fall intentionally. In concentration lower than 0.5 mol / L, the operation of a cell is limited to the current density of less than 100 mA/cm^2. It was found out that the low rate of flow can be applied in the case of low current density. Since it increases but also the transportation mass velocity to the anode of organic fuel not only in order to remove the carbon dioxide generated according to electrochemical reaction when making it operate with high current density, the high rate of flow is required. In the low rate of flow, that fuel passes a film and moves to a cathode from an anode decreases.

Preferably, oxygen or air passes and circulates the cathode 16 under the pressure of the range of 10 - 30psig. It is improved, when a pressure higher than external pressure power is used and the mass transport of wave of oxygen to the place of electrochemical reaction is especially high current density. The water generated by electrochemical reaction in a cathode is carried out of the cathode room 28 by the flow of oxygen through the port 30.

In addition to electric oxidation being carried out with an anode, the liquid fuel which is dissolving in water penetrates the solid polymer electrolyte film 18, and combines it with oxygen on the surface of the electrocatalysis of a cathode. This process mentions methanol as

an example and is expressed with a formula (3). This phenomenon is called "fuel movement (fuel crossover)." Fuel is consumed without the action potential of an oxygen electrode falling and as a result generating useful electrical energy by movement of fuel. Generally, movement of fuel is a parasitism reaction which reduces performance and generates heat within a fuel cell while reducing efficiency.
Therefore, it is desirable to make movement speed of fuel into the minimum. This movement speed is proportional to the transmissivity of the fuel which passes a solid-electrolyte membrane, concentration increases, and it increases as temperature rises. The transmissivity to membranous liquid fuel can be reduced by choosing a solid-electrolyte membrane with low water content. If the transmissivity to fuel is reduced, movement speed will fall. The fuel with a large molecular size has a diffusion coefficient smaller than fuel with a small molecular size. Therefore, transmissivity can be lowered by choosing fuel with a large molecular size. Although water-soluble fuel is desirable, solubility presents transmissivity with low fuel of a medium. Since the fuel of a high boiling point does not evaporate, transportation of these fuel that lets a film pass is performed by the liquid phase. Since steamy transmissivity is higher than a fluid, generally the fuel of a high boiling point has low movement speed. The concentration of liquid fuel can be lowered also in order to make movement speed small. The anode structure over which the hydrophobic site and the hydrophilic part are distributed the optimal prevents an excessive amount of fuel from being able to maintain electrochemical reaction by getting wet moderately with liquid fuel, and approaching a film electrolyte. Therefore, if the structure of an anode is chosen suitably, high performance and desirable low movement speed can be obtained.
Since water can be penetrated at a temperature higher than 60 **, most quantity of water crosses a film by a penetration and evaporation, and a solid-electrolyte membrane is conveyed. The water conveyed through a solid-electrolyte membrane is condensed within a water recovery system, and is sent to a water tank (this water recovery system and tank are explained in detail below with reference to drawing 2), and that water can be made to recycle into the anode room 22.
Inter-electrode [two] is conveyed to the water generated with the proton generated with the anode 14, and the cathode 16 by the proton conductivity solid-electrolyte membrane 18. It is important for operating organicity / air fuel cell effectively to maintain the high proton conductivity of the film 18. The water content of an electrolyte membrane is maintained by carrying out direct contact to liquid fuel and the mixture of water. Let preferably thickness of a proton conductivity solid polymer electrolyte film be the range of 0.05-0.5 mm. If it may become the membrane electrode assembly in which the mechanical strength was inferior when the film thinner than 0.05 mm was used and a film thicker than 0.5 mm is used on the other hand, Superfluous resistance may be presented, while the polymer swells with the solution of liquid

fuel and water, and it is extreme and bringing about the dimensional change which does damage. The ionic conductivity of an electrolyte membrane must be larger than 1-$ohm^{-1}cm^{-1}$, in order to have the internal resistance which can permit a fuel cell.
As mentioned above, the transmissivity of an electrolyte membrane to liquid fuel must be low. Although it was found out that the Nafion (trademark) film is effective as a proton conductivity solid polymer electrolyte film, A film made from perfluoro-** sulfonic acid polymer like Aciplex (trademark) (Japan's Asahi Glass Co. company manufacture) to which Nafion (trademark) and the characteristic are similar, And the polymer membrane which the American Dow Chemical Co. company is manufacturing, for example, XUS13204.10, can be used. The film of polyethylene and polypropylene sulfonic acid, the film of polystyrene sulfonate, and the film (for example, film which U.S. RAI Corporation manufactured) of the sulfonic acid of other poly hydrocarbon bases can also be used according to the temperature and the period when the fuel cell operates. . Acid equivalents differ or chemicals organizations differ. . [whether (for example, the skeleton of an acidic group or polymer is embellished) differs from water content, and] Or low fuel permeability can be attained using the film of the complex which consists of two or more sorts of proton conductivity cation exchange polymer from which the kind and grade of bridge construction differ (for example, the bridge is constructed by multivalent cations, such as $aluminum^{3+}$ and Mg^{2+}).
The film of such a complex can be manufactured and the low penetrable ************ electrochemical stability over high ionic conductivity and liquid fuel can be attained.
It is not necessary to use soluble acid of isolation, or the electrolyte of a base, and a fluid supply type direct oxidation type organicity fuel cell is obtained by using the solid polymer membrane of proton conductivity as an electrolyte so that I may be understood from the above explanation. The only electrolyte is the solid polymer membrane of proton conductivity. Acid of the gestalt of isolation does not exist in the mixture of liquid fuel and water. Since free acid does not exist, the corrosion by acid induction of the cell part article which tends to happen in the organic matter / air fuel cell of the acid base of working art is avoided. This brings considerable pliability to the material selection for a fuel cell and a coordinated subsystem. Since carbonate of fusibility is not formed unlike the fuel cell which contains a potassium hydrate as a liquid electrolyte, battery capacity does not fall. A parasitism short-circuit current is avoided by use of a solid-electrolyte membrane.
Further improvement. The reaction of direct type methanol / fluid supply type fuel cell is as follows.

アノード　$CH_3OH + H_2O = 6H^+ + CO_2 + 6e^-$

カソード　$1.5O_2 + 6H^+ + 6e^- = 4H_2O$

正味　$CH_3OH + 1.5O_2 = CO_2 + 2H_2O$

The various improvement at the time of manufacturing and forming a desirable structure and material which are used by this invention is described by this specification.

It was confirmed by the various experiments conducted by artificers that one specific desirable catalyst material is a platinum ruthenium ("Pt-Ru"). Various processes which combine these two metal are possible. Artificers found out that the direction of the powder of 2 metallicity which has ruthenium particles separate from separate platinum particles brought about a result better than a platinum ruthenium alloy. Since a desirable Pt-Ru material used by this invention has big surface area, it makes contact between this material and fuel easy. Although platinum and a ruthenium were used by catalytic reaction, this artificer found out that it was important that the mixture of a platinum ruthenium is uniformly mixed over the whole material, and the interval is vacated at random, i.e., that material must be homogeneous.

The 1st field of this invention is forming the platinum ruthenium 2 metallic powder which has a unique individual part of a different material combining different metal. Although a certain combination exists among particles, it guarantees that the art of this invention makes the grade of combination the minimum.

The process of forming a desirable material is written in this specification. First, the slurry which put platinum salts and a ruthenium salt into chloride is formed.

Hexahydrate chloroplatinate salt $H2PtCl_6$ and $6H\ _2O$ are made to generate by dissolving a chloroplatinic acid crystal in chloride.

$K2RuCl_5$ and H_2O of a ruthenium salt are made to generate from potassium salt of pentachloro AKUO ruthenium (III).

A 12.672-g chloroplatinic acid crystal is mixed with 600 ml of the crystal of potassium salt of a 13.921-g pentachloro AKUO ruthenium, and 1 mol of chloride. The mixture of acid and a salt is agitated for 15 to 30 minutes, and a homogeneous mixture is obtained.

Subsequently, by adding 140 g per ml/m of sodium carbonate (Na_2CO_3) at the temperature between 20-30 **, it neutralizes to this acid slurry and it is settled. In the meantime, carbon dioxide will be violently emitted from this solution. Adding sodium carbonate is continued until discharge of this gas finishes. That solution changes to a brownish black color at this time. Artificers found out this thing for about 15 minutes to this.

While performing this operation, maintaining to suitable pH is important and pH should be

maintained in the 9.25 neighborhoods by adding sodium carbonate slowly.
Subsequently, this "a gray powdered lump (grey powdery mass)" is processed, and water is evaporated from that slurry. The solid of 1 hour thru/or the shape of black glue which it took 2 hours, and material dried at last, and became a lump is formed in this evaporation. Next, this black glue-like solid is dried in a vacuum, or it is made to dry at 80 ** - 100 ** under a nitrogen air current. A massive gray solid is obtained. The material which still exists in a solution with sodium chloride in addition is included in this solid.
In addition to hydroxylation ruthenium =Ru(OH) $_3$, platinum hydroxide =Pt(OH) $_4$ and "a sticky thing (gunk)", i.e., a chloride, and it, there is superfluous Na_2CO_3 in the chemical contents of this gray powder.
Artificers presume that such excessive materials maintain separation between platinum and a ruthenium. If maintained only at these platinum and the charge of a ruthenium material, they will sinter and join together and the size of particles will become large. The buffer of carbonate between particles has prevented fusion.
Next, it is returned under the atmosphere of hydrogen and argon and this massive solid material changes from the salt to metal. This material is moved to a glass boat and changed. This boat is allocated in the center of the glass tube of a tubular furnace. The material is made to return instead of [its] in the mixed gas which consists of 7% of hydrogen, and 93% of argon near 225 ** in the mixed gas of hydrogen/nitrogen. This gas should have the boat top passed at the per minute 50-200-ml rate of flow.
Maintaining this gas stream is continued by that heated atmosphere for 14 hours. And the catalyst powder is made to cool even per 40 **, still continuing passing hydrogen to the powder. Thereby, platinum, the particles of ruthenium **, and the mixture that consists of other chlorides and carbonate in addition to it are formed.
The obtained material must be washed next. This material performs six washing by several washing, for example, 60 **. By each washing, the sample in a glass boat is moved to the beaker into which 1 l. of 60 ** deionized water was put.
In a platinum ruthenium, it is insoluble to water. Therefore, even if it washes, it is uninfluential in the charge of a platinum ruthenium material, and only other materials are removed. In each washing, since the process of agitating solution for 15 minutes is performed, the chloride and carbonate of fusibility dissolve.
Since the size of metal particles is a submicron, they form the mixture of colloid instead, without sinking to a bottom.
Even 40 ** is made to cool this solution. This solution will be later centrifuged at 3000 rpm for 1 hour. Clear supernatant liquid remains by a centrifugal separation process. It is begun to move the fluid which besides becomes clear, and black sediment is moved and changed into the flask into which 1 l. of 60 ** deionized water was put. Any chlorides which dissolved are

removed by this further washing.

This washing process is repeated 6 times in all. It turned out that it is important to agitate and centrifuge water in order to remove a chloride on the whole. These chlorides are harmful to a catalysis. However, although artificers were binding materials required for these chlorides to suppress fusion of material to the minimum, they found out that it had to be removed later.

After performing final centrifugation operation, the powder is moved to a beaker and it dries in 60 ** vacuum oven for 3 hours. It may replace with this and this material may be freeze-dried.

A platinum ruthenium catalyst [activity / size / which can flow freely / submicron] is acquired by this. It is important to note that a dry material is submicron size, therefore that they are easily carried with air. In order to ensure safety, it must equip with a submicron mask.

It turned out that this activity catalyst powder contains the homogeneous mixture of the platinum particles of submicron size, and ruthenium particles. The residue of RuO_2, ruthenium oxide, and the amount of traces of a ruthenium alloy also exists.

This powder is used as a catalyst on an anode so that it may write in this specification.

The platinum salts and the ruthenium salt which are the first output of this converting method may be changed with a titanium dioxide (TiO2), iridium (Ir), and/or osmium (Os). Such materials can be used that the performance of a fuel cell should be improved at comparatively little cost.

The particles which performed this processing as compared with the particles of the advanced technology show the conspicuous advantage.

The particles of the advanced technology are particles with a size of 5 microns. The ruthenium dioxide was contained in these particles. When the particles of this invention are analyzed, it is shown that it is a homogeneous mixture until it results in the particles of micron size. When it sees with a scanning electron microscope, there is neither a bright point nor a cloudy point, and all the materials look overall to gray. This shows that a material homogeneous on the whole was formed of the mixed process.

The material prepared by this process is called anode catalyst material. It will become "ink (ink)", if this anode catalyst is combined with the Nafion solution etc. and a process is advanced further. That with which platinum metal and a ruthenium metal combined is contained in this so that it may write in this specification. Artificers have found that the desirable ratio to the ruthenium of platinum is between 60/40 and 40/60. It is thought that the best performance is obtained at the time of 60% of platinum and 40% of ruthenium.

Performance will fall slightly, if a catalyst becomes 100% of platinum. Performance will fall rapidly, if a catalyst becomes 100% of ruthenium.

By adding other additives in this salt, the characteristic can be improved and this catalyst material can be replaced by another cheaper material. Artificers thought that this fuel cell should have been formed with the material of a low price. Though regrettable, platinum is a

very expensive material. As long as it is today's writing, a platinum ruthenium is the best material as a catalyst. Artificers have studied using the substitution thing to all or some of platinum of this catalyst. This substitution is due to the point on which a platinum ruthenium catalyst functions.

The reaction which occurs with an anode is CH_3OH+H_2O->CO2+H^++e^- . Artificers think that the catalyst of this reaction is carried out, when a platinum ruthenium carries out the help which makes material dissociate in that catalyst surface. This material pulls out an electron and makes them dissociate. This reaction can be explained as follows.

Methanol is carbon compounds. A carbon atom is combined with other four atoms. three of the bonds come out to a hydrogen atom. another bond comes out to a hydroxyl group, i.e., OH. Platinum dissociates methanol from a bond with the hydrogen, and forms M=C-OH(M is catalyst of Pt or other metal sites)+$3H^+$. A ruthenium dissociates hydrogen from the molecule (HOH) of water, and forms M-OH. The thing in these surfaces ranks second and is newly together put as CO_2+$6H^+$+$6e^-$. An anode generates H^+ (proton), it is crossed from the anode to a cathode, and is returned there. This is called 2 functionality catalyst.

Methanol which was written in this specification, and any materials with the similar function to make water dissociate can be used instead of platinum. Artificers studied several sorts of such materials. They found out the substitute of platinum, such as palladium and tungsten which can dissociate a C-H bond, rhodium, iron, cobalt, and nickel. It seems that molybdenum (MoO_3), a niobium (Nb_2O_5), a zirconium (ZrO_2), and rhodium (Rh) have the performance which dissociates and makes H-OH M-OH. Therefore, it is the good catalyst which combined these. Ru, Ti, Os, Ir, Cr, and/or Mn are mentioned to the desirable catalyst which dissociates a H-O-H bond.

The material which all resembled the ruthenium in the part may replace a ruthenium. Artificers found out having many characteristics that iridium resembled the ruthenium. Therefore, in the 1st embodiment in this field, what combined platinum, a ruthenium, and iridium by the relative ratio 50-25-25 is used. This adds salt H_2IrCl_6 and H_2O in the quantity which was suitable for the first material indicated above considering it as the combination of 50-25-25 (Pt-Ru-Ir).

It turned out that fewer rutheniums are used for this catalyst and it works very well.

The material containing a titanium compound is mentioned as another material in which it turned out that it has a certain advantage. Any titanium alkoxides and titanium butoxide, for example, titanium isopropoxide, or $TiCl_4$ can be added into the original mixture. Of this, the final combination of platinum-ruthenium TiO_2 can be formed and it is also formed in the combination of 50-25-25 (Pt-Ru-TiO_2).

Platinum-ruthenium osmium can also be used. Osmium was added by the mixture as salt H_2OsCl_6and6H_2O, and it became clear that the advantageous characteristic was brought about by this.

Even when formed how, such materials used in order to form platinum ink must be attached to an anode. Various art can be used in order to attach this material. Then, formation of an anode is explained below.

Formation of carbon paper. Movement (crossover) of fuel caused degradation in this fuel cell. Instead of reacting with an anode, methanol happens, when fuel movement in this fuel cell passes through an anode. Methanol passes through an anode and penetrates a membrane electrode assembly, a film, and a cathode. Methanol may react to a cathode, and the efficiency of fuel will be reduced if it becomes so.

The electrode of this invention is preferably formed using the base of carbon paper. Start material is TGPH-090 carbon paper which can be obtained from Toray of the New York State City of New York third avenue 500 whereabouts. However, this paper pretreats first and improves that characteristic. The "Teflon 30" suspension of DuPont of about 60% of solid is used for this pretreatment.

This paper may be a carbon fiber of the fragment mixed with the binder. After carrying out the roller cliff of this fiber, it forms the last material of about 75% of porosity by carrying out combustion removing of the binder. It is also possible to use a carbon crossing paper. This is processed by the art written in this specification. Carbon paper crossing can also be used. This is processed by the art indicated here and forms the backing of gas diffusion nature / current collector.

The carbon paper carried out in desirable processing contains the paper with which Teflon particles were embedded inside. As for the space between Teflon particles, it is preferred that it is sufficiently small so that methanol cannot pass through that. For example, the better characteristic is used even when using other methanol derivatives like TMM. This anode assembly is formed on the base of carbon paper. It means that this carbon paper is Teflon-ized, it adds Teflon, and the characteristic is improved. Artificers found out that there was balance important between the quantity of the Teflon added on a paper and the final characteristic.

It is important to maintain the quantity of the Teflon to be used to suitable balance so that it may write in this specification.

While Teflon-izing this paper and giving water repellence, the mixture of platinum ink is kept from oozing through that paper.

Although this paper needs to be wettability, it does not need to be porosity. This delicate balance is brought about by immersing and heating a paper. Since artificers found out that balance existed between the grade and the amount of being impregnated to a paper of the

wettability of a paper, they explain that here.
First, the emulsion of Teflon 30 must be diluted. Every 1 g of Teflon 30 is added in the water in every 17.1 g. 1 g of Teflon [60% of the weight of] 30 is equivalent to 60 g per 100 ml of Teflon.
This material is poured into a suitable container like a glass dish, for example. Carbon paper is maintained until it sinks in into the material.
Immersion operation carries out the amount of criticism of the carbon paper of one sheet, and subsequently to the process of dipping carbon paper, for about 10 seconds, it corresponds in the solution until it gets wet clearly. Carbon paper is picked out from a solution using tweezers, making it not contact the paper as much as possible. However, the tweezers itself draw the Teflon on the character of Teflon, and distribution of a fluid becomes uneven. In order to make this possibility into the minimum, the tweezers by which the Teflon coat was carried out are used. Carbon paper turns an angle downward, is held and makes a superfluous solution flow out.
Many Teflons will be torn off by surface tension if the material lays to a glass surface with the surface tension characteristic of a Teflon emulsion. As shown in drawing 3, a paper dry assembly is formed without doing so.
The wire 202 which gave two or more Teflon covering is stretched on a perforated plate like the dish 200. This stretched wire forms the two-set base materials 202 and 204 intersected perpendicularly and prolonged. The carbon paper just processed with the Teflon solution is held ranging over these base materials.
Ideally, these wires are wires 0.43 inch in diameter which carried out Teflon covering. Although these sizes are not delicate, if the area in contact with a paper is small, distribution of the suspension on a wire can be made more into homogeneity. The kink 206 is formed in the wire, and carbon paper is kept from contacting a wire along with the overall length of the length direction, and makes the touch area still smaller by it.
Subsequently it is put into the paper dry assembly shown in drawing 3 into 70 ** oven for 1 hour. The carbon paper which processed is removed from a dish after desiccation, and is put in glassware. Then, these are sintered within a 360 ** oven furnace for 1 hour. Weight is increasing the paper processed appropriately by 5% through this down stream processing. It is permissible if weight increase's being for 3 to 20%, speaking still more generally. ******** [that absorption with this sufficient paper was made] -- and/or, in order to decide whether it is necessary to process a paper further, the amount of criticism is carried out.
This substrate + catalyst bed forms a final electrode.
Two desirable art which attaches the catalyst containing a layer is explained here. That is, they are the direct applying method and weld slag vacuum deposition. Other carbon paper containing the carbon paper used without using a special carbon paper material which both

explained the method of formation of law to the above and performing special processing in any way may be used. The charge of a platinum ruthenium material and the various materials which explained the method of applying this invention above directly are mixed, or a catalyst material and various materials are mixed on other combination drugs of a certain and a general twist target. This catalyst material is processed with the material of the addition which improves the characteristic.

It mixes with an ionomer and platinum ruthenium powder is mixed with water repellent. Perfluorosulfonic acid (Nafion) and the solution of the micro particles of Teflon are mentioned to a desirable material. 5 g [per 100 ml of Nafion solutions] platinum ruthenium powder is added.

T-30 mixture of DuPont of 60% of the weight of the Teflon solid diluted suitably is added. Then, the micro particles of such Teflons are mixed. Dilution Teflon 30 suspension which contains in 4 g of deionized water preferably the solid of 12 mass percents into which the concentrate of 1 g of Teflon 30 was put is prepared. 300 mg of deionized water is added in 12% of the weight of the above-mentioned Teflon solutions [350 mg of]. A 144-mg platinum ruthenium is mixed in this solution. Subsequently the obtained mixture is mixed using a publicly known ultrasonic blending technique by conventional technology as "sonication." Ultrasonic mixing is preferably performed within the ultrasonic bath which filled water with Fukashi (about 1 / 4 inches). It is carried out for [of this mixture] about 4 minutes "ultrasonication."

As mentioned above, the point that Teflon must be first mixed with a platinum ruthenium and about 15% of the weight of Teflon must be prepared is important. Nafion can be added only after making this mixture. Artificers found out that it might enclose the particles of platinum and a ruthenium, if Nafion was added first. Therefore, an order of this operation is delicate and important. At this time, 5% of the weight of Nafion [0.72 g of] is put into a jar, and sonication is performed for 4 minutes once again. 1 square centimeter of electrodes covered as more generally saying hit, and it is necessary to add 1-mg Nafion about. The quantity of above-mentioned Nafion is also slight, or when only 652 ml adds, it can change a solution, for example.

If this processing is performed, a black material of slurry form will generate. Next, the black material of this slurry form is applied to carbon paper. The applying method may be performed by one [which] of many methods. The easiest gestalt is applying the material on carbon paper backing, changing a stroke in the different direction. In order to apply this, a brush with small hair of a camel is used. A desirable quantity which material mentioned above is the quantity for forming the catalyst of sufficient quantity to one side of 2 inches x the piece of 2 inch of 5% of the weight of Teflon-ized carbon paper. Therefore, spreading is continued until it will use all the catalysts.

The drying time for 2 minutes - 5 minutes should be established between covering, and each

covering should be applied in the different direction so that material may be in a semiarid condition between covering. It is necessary to dry an anode for about 30 minutes after that. "Press working of sheet metal" of the anode must be promptly carried out after progress of the 30 minutes. Operation of this press working of sheet metal is explained in this specification.
The obtained structure is a porous carbon board used for diffusing gas and a fluid.
It is covered with 4 times per 1 square centimeter of catalyst materials.

The alternative technology to which this material is made to adhere is carrying out the weld slag of that material on backing.
We have explained how to form an anode here. Next, it ranks second about the art in which the process of forming the film (Nafion) of desirable proton conductivity involves, and the art which forms a cathode is explained.
Proton conductive film - A desirable material indicated here is Nafion 117. However, in order that other materials may form a proton conductive film, it can use. For example, it is possible to use other perfluorosulfonic acid materials. It can be assumed that many different materials with a carboxylic acid group may also be used for this purpose.
A desirable embodiment starts using Nafion 117 which can be obtained from DuPont. This material is first cut in a suitable size. It is important to use a suitable size and it is because the conditions of the last material are decided by the size. First, Nafion is boiled in a hydrogen peroxide solution. 5% of hydrogen peroxide solution is got, and this film is boiled at 80-90 ** in this solution for 1 hour.
Oxidizable organic impurities are removed by this.
After performing the boiling step by this peroxide, that film is boiled for 30 minutes at about 100 ** in deionized water. The hydrogen peroxide absorbed by the film before that is removed with other water-soluble organic materials.
Thus, the film to which processing was performed is boiled in a sulfuric acid solution next. A 1-mol sulfuric acid solution is prepared by diluting sulfuric acid of the condensed 18-mol commercial ACS grade. Acid of the ACS grade contains only the metal impurity of a quantity smaller than 50 ppm. By boiling this film at about 100 ** in 1 mol of sulfuric acid, that film can be more nearly thoroughly changed into proton conductivity.
The processed material is boiled for 30 minutes at 90-100 ** in deionized water next.
That water is thrown away, this boiling step is repeated further 3 times, and membranous purity is raised.
After performing these washing processes, sulfuric acid does not exist in the film, but it becomes a "protonic" gestalt thoroughly. It is put in and saved by that in the sealed container at deionized water until the film is ready to perform further processing.
Cathode composition. A cathode is constituted by preparing the catalyst ink for cathodes first.

Preferably, although the catalyst ink for cathodes is pure platinum, as other ink can be used and being written in this specification, it may mix many of other materials in ink. A 250-mg platinum catalyst is mixed with 0.5 g of water containing 37-1 / 2 mg of Teflon. Sonication of this mixture is carried out for 5 minutes, and it is aligned with 5% of Nafion solution. Sonication of the obtained mixture is again carried out for 5 minutes, and a uniform dispersing agent is obtained. Sufficient material to cover the 2x2-inch piece of carbon paper with this is formed. Unsettled Toray carbon paper can be used without Teflon content. However, preferably, this material is Teflon-ized, as mentioned above.

Subsequently, the process of forming the paper with which 5% of Teflon was impregnated continues. Then, the paper is heated at 300 ** for 1 hour, and Teflon particles are sintered. The material is covered with PT of 4 $mg/cm^2/g$ by applying after that, as catalyst ink was mentioned above on the paper. Although the Teflon content of this paper may change among 3 to 20%, it is desirable. [5% of]

In the alternative technology which forms sputtering cathode, the platinum electrode which carried out weld slag formation is formed.

It became clear that it had the advantage conspicuous when this platinum electrode that carried out weld slag formation was used as a planate air electrode. The process of manufacturing the platinum electrode of sputtering formation is written in this specification.

A cathode terminal performs a reaction called $O_2+H^++e^-$ -> water. This O_2 is received from the surrounding gas (gas) of a platinum electrode, and an electron and a proton are received via a film to this. This alternative technology for forming a cathode terminal starts using platinum of the grade for fuel cells. This can be purchased from many sauce, such as Johnson-Matthey. 20-30 g per surface area of 1 square meter of this platinum is applied to that electrode in the size of a 0.1-1-micron particle.

The sauce of platinum is solid-rods-like material. According to this embodiment, the weld slag of that material is carried out to the substrate formed as mentioned above. Platinum powder is first mixed with an aluminium powder. This mixing may be performed using the art of salt combination which was performed when it could carry out, for example using a mechanical means or Anode, Inc. was manufactured and which was mentioned above. The weld slag of the platinum aluminum mixture is carried out on carbon paper using a known sputtering technique by semiconductor technology.

The weld slag of the platinum is carried out as follows using the system illustrated by drawing 4. The carbon paper electrode 252 is held at the 4-inch standard target 250. This target rotates one time in 10 seconds by the motor 254. According to the desirable art of using here, the weld slag of the Pt is carried out from 1st Pt supply source, and the weld slag of the aluminum is carried out from the aluminum supply source 262. Near 200 volt, platinum is 0.23amps and the

weld slag of the aluminum is carried out by 0.15amps. Two supply sources collide at the angle of 45 degrees toward a target from a different direction which faced each other.
Although the pressure with artificers ideal for this sputtering was 20 torr, it found out that the arbitrary pressures between 1-50 torr could also be used. The pressure of argon is an about 30-mm torr. However, the atmospheric pressure of argon which is different in order to form the particles of different size can be used. Sputtering is performed for about 8 minutes.
Preferably, after finishing sputtering, by immersing the backing which carried out weld slag formation in an etching solution, it etches, and it is continuously immersed in a washing solution.
The electrode which carried out weld slag formation is a mixture of the Al particles on backing, and Pt particles. An aluminum particle is removed by washing an electrode with a potassium hydrate (KOH). Of this, the carbon paper backing to which porous platinum adhered dramatically is formed. Each field in which aluminum was formed is removed and the space which the hole opened to the position remains. When artificers' covering of Pt-Al material was thick, that aluminum is flushed from a downward field of the catalyst found out being prevented a little. This invention uses thin covering (coating), it is a desirable tunic in which it is thinner than 0.1 micron or it, and material density is between cm^2 the whole 0.5 mg with cm^2 the whole 0.2 mg.
The anode, the film, and the cathode were made in this this time kicked in this process. Such materials are assembled by the membrane electrode assembly ("MEA").
the formation electrode and film of MEA -- a 5Mil thickness [of the CP class], and titanium foil (12-inch x 12 inches) top -- first -- arrangement -- namely, -- carrying out a stack (pile) -- **.
Artificers use titanium foil so that any acid components which come out from a film may not permeate foil.
First, an anode electrode is placed on the foil. That the characteristic of a desired film should be maintained, it soaks and the proton conductive film is saved. This proton conductive film is wiped first, and is dried, and particles with large size are removed. And the film is directly placed on an anode. A cathode is placed on the film. The cathode is covered and another titanium foil is allocated.
The end of the titanium foil of two sheets is stopped with a clip together, and the layer of material is held to a position. The titanium foil and the film for pressing on both sides of an assembly in between contain the stainless steel plates of two sheets about 0.25 inch thick, respectively.
The film and electrode in the titanium foil assembly stopped with a clip are carefully allocated between the stainless steel plates of two sheets. These two plates are held, for example between the jaws of a press device like an arbor press. The press device should be maintained by low temperature at the room temperature, for example.

Then, the press device is operated and the pressure between 1000 to 1500 psi and the optimal pressure generate 1250 psi. The pressure is maintained for 10 minutes. Heating is started after for these 10 minutes passes. Although heating is slowly raised to about 146 degrees, it turns out at any temperature in a 140-150 ** temperature requirement that it is effective. In order to raise temperature slowly, many hours should be spent for 25 to 30 minutes, and it is the time for stabilizing temperature for 5 minutes of the last of heating. The temperature is stopped at 146 ** for about 1 minute. The pressure is maintained although heating is stopped at the time.

Then, the press device is quickly cooled using circulating water, maintaining a pressure to 1250 psi. The pressure will be opened, if about 15 minutes pass and temperature amounts to 45 **. Then, the film and electrode which were combined are removed and it saves in deionized water.

Flow field. The fuel cell operates appropriately, only when fuel is appropriately supplied to the film so that a reaction may occur or a catalysis may occur. The membrane electrode assembly of this invention uses a flow field assembly as shown in drawing 5. Each membrane electrode assembly ("MEA") 302 is inserted among the plates 304 and 312 for flow change of the couple which has a by plate and an end plate. The flow of fuel is established to each space 303 between each by plate / end plate, and MEA. A by plate / end plate, and MEA(s) gather, and a "stack" is formed. The by plate equips with the composition for the flow of a fluid both the surfaces that face face to face. The flow plate of the end of this stack is not a by plate but the end plate 312. The end plate has a chamber only in one side. Two or more separators 306 and two or more chamber formation areas 308 are established in the by plate 304. The separator 306 has a function forced to the membrane electrode assembly 302. The end face of the separator 306 is the planate surface substantially.

It contacts the surface of MEA302.

The by plate is formed with the conductive material so that all the membrane electrode assemblies 302,310 may be combined in series mutually.

The membrane electrode assembly 302 which was mentioned above is provided with the anode, the film, and the cathode. Anode side 312 of each membrane electrode assembly touches the supply source of aquosity methanol in the space 314. The cathode side of each membrane electrode assembly touches the source 316 of oxidizer air supply which provides the gaseous substance for performing the reaction explained above. This air may be mere air and may be oxygen.

The flow of these charges of green wood is required in order to continue supplying fuel to an electrode appropriately. It is also desirable to maintain the stable flow.

An example of a stack design of this invention uses the system shown in drawing 6.

Fuel is supplied from the fuel supply chamber 602, and it is the room of high capacity into which the pressurized fuel usually went. The portion 604 like a narrow nozzle reduces a pressure greatly along with it. The fall of the pressure in this thin pipe is much larger than what kind of the failure of pressure along a supply route. The flow between the cells within a cell is equalized by this.
The careful balance of balance must be taken between the total amount of the surface which acts as an application-of-pressure element, and the total amount of the surface which acts as a holding element.
It is desirable to put a uniform pressure on the membrane electrode assembly 202 for various reasons from both sides. However, when the application-of-pressure surface 306 is pushing to the film, direct contact of the membrane electrode assembly 302 cannot be carried out to methanol. Instead, it is pushed there by the surface 315. Therefore, the portion of the surface of MEA302 does not react. It has various functions, and the thing of the various designs by this invention improves a flow, or improves some characteristics in the field of reliability.
The width of each nozzle 606 is narrow. The exit 605 of each nozzle 606 faces one island which pressurizes the field 608 corresponding to the application-of-pressure surface 306. Direct supply of the fuel supplied from the nozzle 606 is carried out to the interface 610 of the island 608. The island shown in drawing 6 is a rectangle. The interface 610 is a side with the narrower island of this rectangle. The side with this larger island is parallel to a flow. All the input flow goes to one of the surfaces of an island directly.
Artificers found out that a turbulent flow was formed in the field of the island 608 according to this desirable narrow layout. A turbulent flow agitates the fuel in a chamber and stabilizes more the flow which passes along this system. This turbulent flow makes the flow between each island easy. An output flow is eventually accepted by the output nozzle 612, and is led to the output tube 614. An output nozzle is allocated in the form which adjoined on the surface of [620] the island, and was similar, and a turbulent flow is further formed of this.
Side 610 of a boundary is 50 mils and the larger one side of the island by this embodiment is 150 mils. The failure of pressure in the both ends of a stack is about 0.06 psi about this stack.
The by plate of other allocation configurations can also be used.
The by plate itself is lightweight, it is thin, and it is important for it that the interval of a stack can be enlarged as much as possible.
Graphite is difficult for machining it and it is comparatively thick. Passing neither a fluid nor a gas is mentioned as an advantage of graphite.
According to this invention, many another solutions are used. Change of the 1st of this invention uses a system as shown in drawing 7.
the boundary layer 702 -- high -- it is dense and is a layer which does not let a conductive fluid or gas pass. By this, the quantity of the fuel which can cross a wrap by plate assembly for

material, a gas, or a fluid is decreased. However, a dense porous material is used as the crossed area 700. A certain amount of quantity of material will be infiltrated into MEA through this boundary layer as it is porosity.
A dense porous material may be conductive carbon machining is much more [than graphite] apt to carry out, for example. Although osmosis is dammed up by boundary material, it can prevent a fluid and a gas from crossing the whole by plate by this.
If the pressurized part has porosity, a fluid and a gas will be made to reach a certain amount of portion of the membrane electrode assembly currently pressurized by the pressurized part. Therefore, methanol may permeate these fields and conversion efficiency will decrease in the another state.
The main binder layer 704 is carbon of low density ("LD"). It is comparatively easy to deal with LD carbon, and its price is also cheap. However, since all the portions are covered by graphite as for LD carbon, the fact, most of the characteristic which is not desirable, is hidden.
The 2nd embodiment used by formation of a by plate is shown in drawing 8.
The by plate of this 2nd embodiment is an ultra thin type by plate made from titanium carbon which became stratified. Since we desire a stack made as thin as possible to arbitrary request voltage, a by plate must be thin. If each membrane electrode assembly and by plate are activated, voltage will occur, but it is called peculiar voltage (inherent voltage). Peculiar voltage and the thickness of this device determine the greatest voltage that can be taken per inch in thickness of the device of this invention. The important element about the voltage to thickness is the thickness of a by plate.
The by plate of the 2nd embodiment of this invention is shown in drawing 8. The thought of the stratification of forming a by plate combining the best characteristic of that material is used for this material. The boundary layer 800 of titanium carbide is joined to the titanium bond layer 802. The titanium bond layer 802 is preferred and thickness is 3 mils. These two layers became together, and suitable electrical coupling nature has also secured them while they prevent that cross a by plate and a proton moves. This titanium material is covered with the charge 804 of a separating material, and holds a by plate to a prescribed position. Therefore, a certain amount of porosity is possible similarly in the embodiment of drawing 7.
Although it is natural, titanium can replace any metal with the same conductivity and chemical stability.
The graphite material used ordinarily recognizes the artificers of this invention that the balance between the claims postulated which compete will surely arise.
In order to improve operating efficiency, it is required for fuel to cross to the interlayer side who faces the same another by plate side, i.e., a cathode, and to keep it from sinking in from the one, for example, anode, one by plate side. Supposing a by plate is porosity, fuel material may be crossed and it may sink into a loan. However, since there is no fluid which can pass a by

plate, it means that there is no fluid which arrives at the field of the electrode layer assembly currently pressurized by 306, for example with a by plate with the application-of-pressure surface. Therefore, in these portions currently pressurized by the application-of-pressure surface in these membrane electrode assemblies, electrical activity does not occur efficiently. The efficiency of this whole cell worsens by this.

In these embodiments of this invention, a new kind of balance matter (matter of merits and demerits) occurs. A membrane electrode assembly is pressurized by the porous portion of a by plate. That fuel comes to be supplied a little to the portion by which the electrode was pressurized at least by having used this porous portion. By this, the electrical performance nature of MEA is improvable. According to this feature of this invention, other connecting pieces which prevent that a fluid passes into other portions of an electrode layer assembly can be provided.

System. The fundamental system of this invention is shown in drawing 9. This system is based on the way of recycling the output of a fuel cell which artificers have recognized.

A fuel cell consumes methanol, a methanol derivative, and water, and output output, such as methanol or a methanol derivative, water, and gas, generates it. Methanol means the fuel consumed. Any fuel cell systems must carry the methanol fuel of quantity consumed. However, the reaction also needs equivalent weight of water. Artificers have noticed that it can make the water used at this reaction collect and recycle from a cathode. By this, the necessity of carrying a lot of water is avoidable.

A work load generable from a vehicle is restricted by the gross vehicle weight (pay load), i.e., the weight of a vehicle and its mounted object. Every vehicle has a limit in power with the weight which it must carry. If weight is large, the power of a vehicle will be restricted, therefore the efficiency of a vehicle will become small. For example, the passenger car has not usually carried gasoline of 20-30 gallons or more. This is decided from things various so that between the distance which can run by the time a vehicle re-refuels a tank, and the weight which increases by enlarging a fuel tank may serve as optimal balance.

The engineer of a vehicle determines how much a pleasantly permissible pay load is. Artificers explain the art of securing this pay load being taken not with water but with fuel.

One of the features of the system of this invention is maintaining the balance of water, as most water is recycled and it does not have a substantial supply source of the water which needs to be carried.

The whole system is shown in drawing 9. The methanol tank 900 saves pure methanol (or fuel of a derivative methanol type [other]). The 1st liquid pump 902 carries out pumping of the methanol to the circulation tank 906 through the valve 904. The water tank 908 supplies water to a required place. Pumping of this water is carried out to the recycle tank 906 through the valve 912 with the pump 910. The prime controller 914 controls the synthetic operation of this

whole system. The control device 914 controls the relative position of the valves 904 and 912. The methanol concentration sensor 916 has a preferred thing of the inside of methanol, or methanol attached to the place of a paddle gap very closely. The methanol sensor 916 detects the methanol concentration in a circulation tank, and the control device 914 controls the operation of this system further based on this information.
Aquosity methanol included in a circulation tank is maintained by 1-2M with this control system. Therefore, methanol in the line 918 should also have suitable concentration. The pump 920 carries out pumping of the methanol to the membrane electrode stack 924 through the fuel filter 922. The stack used here should just be the same stack as what was mentioned above. The electric output 926 which comes out of the stack 924 operates other electric systems like the control device 914 930, for example, a compression equipment, while it is sent to a motor and moves loading load.
This stack operates using the incorporation air 932 which passes along the compression equipment 930. The air filter 934 defecates air, before going into a stack.
The fuel outlet of this stack contains two ingredients, i.e., water, and methanol. Both methanol and water can be condensed by processing two ingredients using the condensing plant 940 and 942, respectively, and making temperature of water low enough. The fan 944 can be used in order to perform this cooling easily. As for methanol and water which are recycled, both are returned to a circulation tank. The methanol 946 recycled from the exit of a methanol stack, the air recycled from the incorporation air 952, and water are collected to the circulation tank 906. It is dramatically uneconomical that a fluid engineer does pumping of the gas, if it sees from a point of the source of energy supply, but carrying out pumping of the fluid to this knows that expense will not increase so much. In respect of there being this invention, the cathode will be required to pressurize air. For example, it may be required to pressurize air at 20 psi. However, the air (after reacting with a cathode) on the line 944 which came out is compressed in most cases highly. Probably, this output air 944 is compressed into 19 psi. In this way, the output air 944 goes into the pressure drive type turbine 946. This expander device operates with a pressure, and since the air compression equipment 930 is driven, it is used. If the compressed power is not reused in this way, 20 to 30% of the electric power generated with the fuel cell may be used for air compression equipment.
The output 948 of the expander device includes the combination of air and water. This water and air are separated and the water which discharged air was breathed out and collected by 950 is returned to the circulation tank 902. Probably, the water flowing hole 954 which drains superfluous water is also needed.
This water flowing hole is controlled by the control device 914, and when there is too much water which is re-flowing back, it is sometimes needed.
The quantity of the fuel supplied may be measured as a means replaced with a sensor and a

control device. However, the latest fuel cell is planned so that it may operate at about 90 **. The reaction of an electrochemical fuel cell makes even a suitable temperature heat the fuel cell after all.

This invention operates using a methanol sensor. Especially the desirable methanol sensor uses the MEA art mentioned above. As mentioned above, a fuel cell comprises an anode and a cathode. An anode receives methanol. A cathode receives air or oxygen.

The fuel cell which was shown in drawing 10 and which changed is used for this sensor. The Pt-Ru anode 1002 is joined to the Nafion electrolyte 1004.

The electrolyte is joined to Pt cathode.

The larger one of this cathode than an anode is preferred, for example, the area of a cathode is 3 times the anode.

The cathode 1006 (and anode) is immersed in methanol solution. Therefore, the reaction from which H_2 is set to H_2O so that it cannot react to air since the cathode 1006 is in a fluid, therefore it may happen by a fundamental fuel cell reaction cannot occur. If voltage is applied to this fuel cell, the occurring reaction will change, for example, will be reversed. When current occurs, an anode reacts to methanol directly, and CO_2 is generated, and a cathode changes a proton to hydrogen. If a cathode is small and an anode is large, a proton will be returned and the susceptibility of this methanol electrode will be improved further.

Therefore, this reaction contains each of following formulas.

(+) H_2O+CH_3->CO_2+ $6H^+$+ $6e^-$ (-) $2H^+$+ A $2e^-$ ->H_2 constant voltage is applied by the constant voltage circuit 1010. The ammeter 1012 measures current. Drawing 11 shows the relation of the methanol concentration in current and a solution. The control device 1014 should just be a process controller or a microprocessor, and carries out the lookup of the nearest methanol concentration corresponding to the current measured using the relation by which drawing 11 was plotted.

Since it seems that temperature dependence is large, the plot of drawing 11 is good to use a thermo couple for correction for temperature.

****** -- another important feature of an invention is related with actually using this system in the environment of a car. It will be necessary to supply methanol with the methanol device of the gas pump, in order to use it practical. The impurity of hydrocarbon may be included when picking out methanol from a gas pump. If there is such an impurity, it is dramatically dangerous with the system indicated by this invention that needs methanol with high purity. Therefore, a fuel filter is used in this invention. This fuel filter is illustrated by drawing 12. The filter of the three-stage includes the zeolite crystal 25M (Mobil) composite type or neutral type. Usually, zeolite acts as a molecular sieve. This zeolite crystal is used in order to filter methanol and to remove a certain hydrocarbon impurity from there. These zeolite is good to include the one-set

layer system which consists of three or more layers with the hole of a size which is different gradually to the layers 1-3 in 3-10 A.
1 2 3 CH_3 OH, H_2O, O_2, and H two-layer 1 are the zeolite X with the hole of a usually big diameter, an offer light, and A, and can remove a big molecule. Mordenite and natural zeolite are used in the layer 2, and it removes n-paraffin, n-butane, and n-alkanes. The zeolite 3A and 4A can be used in order to remove a small molecule like propane or ethane in the layer 3. This is preferred when the molecular sieve which changes gradually is formed.
Unipolar approach.
In the approach of before about a fuel cell, many fuel cells were used in series. If a fuel cell is connected in series, it is possible to make the whole voltage high so that output voltage can be added and the output of a stack may be made to increase to high and more nearly available voltage. The artificers of this invention have noticed that the effect of a seed that this ******** has used unipolarity and the approach which attached the name is acquired further, using the approach which has not been made into the stack. The approach of this unipolarity is maintaining each membrane electrode assembly in the state where it dissociated thoroughly [other components of all the]. According to this approach separated thoroughly, each element of an assembly can have better efficiency while being made much more greatly. The element of each unipolarity may be assembled to a stack. The important things to depend on this feature are that each membrane electrode assembly is connected separately and that that element connected separately is not assembled by the stack and connected in series.
The 1st embodiment of the unipolar invention is shown in drawing 13. This mode can be used in order to form the fuel cell which does not need contact force in order to form electric connection. The film 1300 is preferred in it being the Nafion film. The Nafion film has the piece 1302 of a metal cross, for example, the central region in which the end of the screen was established. The metal cross or the screen 1302 is covered with the suitable catalyst of the type mentioned above. With the tab 1304 which current carries, the generated voltage is connected outside.
As for the flow field insert part 1306 of the product made from a plastic, or metal, a suitable fuel material is led to each side of the crossing which gave the catalyst tunic. The flow field member 1308 may be formed in the side of another side.
Therefore, by attaching to Nafion backing the material which provided the catalyst in the surface, and pressurizing toward it, although it is electrically the same, a fuel cell can be formed by a mechanically different method.
Drawing 14 shows the sectional view of this device. The tab 1304 sends current through the electrode region 1400. Methanol is sent to the methanol chamber 1402, i.e., the sealed field by the side of [one] membranous. This sealing is maintained by the ring shape sealed area 1406. Air is led to the 2nd membranous side through the air chamber 1408 sealed similarly in

an opposite hand. Each of these components operates as a unit of a standalone version regardless of other units. Higher voltage can be provided by connecting in series the current generated from these components.
The 2nd alternative embodiment of this invention is shown in drawing 15. The film 1500 which met the titanium sheet 1502 is used for this mode. Spot welding of the titanium crossing 1504 is carried out to the titanium sheet. The titanium crossing 1504 functions as a cathode and may be covered with platinum. The titanium crossing 1506 functions as an anode and may be covered with the suitable platinum ruthenium.
A gasket and the ring 1508 for combination form the chamber 1510 between a film and an anode. Similarly, another gasket and ring 1510 for combination form a chamber between a film and a cathode.
The titanium sheet equips the surface with the spherical seal 1512, in order to maintain a chamber. The voltage generated from this titanium sheet is combined with the extraction field 1514 of current.
Since metal fasteners can be used if a bead sealing is used, in this mode, the place to which a rivet or a fastener is attached is also provided.
Especially this system integrated can be made very thin if titanium foil is used.
This embodiment can be used with the device shown by drawing 16. The cathode and the anode are provided in each of these double battery modules shown in drawing 13 or 15. The component shown in drawing 16 is assembled so that two anodes may adjoin.
The cells 1602 and 1604 which face mutually are completed.
The flow field 1606 is formed among the anodes 1602 and 1604. This flow field must contain the air which flows between them. Similarly, it faces mutually, the flow field 1608 was formed among them, and the flow of suitable air has produced two adjoining cathodes there.
Drawing 17 is an enlarged drawing about the method by which these cells are used. The flow field 1700 is the flow field of air facing cathode side 1702 of the 1st cell 1704. Anode side 1706 faces the flow field 1708 of the 2nd methanol. Methanol flows from the methanol input 1710 and flows out from the tap hole 1712. The flow field of this methanol faces anode side 1714 of the 2nd bipolar battery 1716. Cathode side 1720 of this 2nd bipolar battery 1716 faces the portion 1722 of another flow field through which air flows.
Although only the embodiment of a mere small number was explained to the above in detail, to be sure, the person skilled in the art can understand that various changes can be carried out to a desirable embodiment, without deviating from this instruction range.
It is interpreted as such all changes being included by the following claim.

[Translation done.]